

Magnetic-Field-Induced Crossover to a Nonuniversal Regime in a Kondo Dot

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Abstract

We have measured the magnetic splitting, Δ_K , of a Kondo peak in the differential conductance of a Single-Electron Transistor while tuning the Kondo temperature, T_K , along two different paths in the parameter space: varying the dot-lead coupling at a constant dot energy, and vice versa. At a high magnetic field, B , the changes of Δ_K with T_K along the two paths have opposite signs, indicating that Δ_K is not a universal function of T_K . At low B , we observe a decrease in Δ_K with T_K along both paths, in agreement with theoretical predictions. Furthermore, we find $\Delta_K/\Delta < 1$ at low B and $\Delta_K/\Delta > 1$ at high B , where Δ is the Zeeman energy of the bare spin, in the same system.

Universality and scaling [1] describe many phenomena, both equilibrium (e.g. thermodynamics of near critical fluids or ferromagnets) and non-equilibrium (transport, turbulent flow). A famous quantum system exhibiting universal dependence of properties on a single intrinsic energy scale (called the Kondo temperature, T_K) is the Kondo singlet, a correlated ground state of a confined spin interacting coherently with delocalized electrons [2]. While the equilibrium scaling in Kondo systems is now well understood, a lot less is known about the nonequilibrium regime, i.e. when the Fermi sea near the spin confinement site is perturbed. The nonequilibrium Kondo phenomena, governed by the delicate interplay of correlations, spin coherence, and dissipation, can be observed in Single Electron Transistors (SETs) [3, 4, 5, 6, 7]: electric transport in SETs is strongly affected by the Kondo effect, and the deviation from equilibrium near the spin confinement site (called quantum dot) can be precisely controlled by an externally supplied bias voltage, V_{ds} . One of the open questions is whether the nonequilibrium Kondo physics is universal, and, if so, whether T_K , as defined for an equilibrium system, remains the relevant energy scale. Very recently, Grobis et. al. [8] have shown that, at low biases and temperatures, the SET conductance $G = \partial I / \partial V_{ds}$ is a universal function of $eV_{ds}/k_B T_K$, where e is the electronic charge. The splitting of the zero-bias Kondo peak in $G(V_{ds})$ in an external magnetic field B [9] is another nonequilibrium effect predicted to exhibit universal dependence on T_K , with B/T_K being the scaling variable [10, 11]. These predictions have not been systematically tested in reported studies [3, 4, 12, 13, 14, 15, 16].

In this Letter, we investigate the dependence of the Kondo peak splitting on T_K at different B by employing two independent parameters to tune T_K , rather than a single gate voltage [15]. This new approach tests whether the changes in the splitting with T_K have a universal character at each B independent of the observations at other values of B . An attractive feature of this method is that a weak dependence of T_K on B due to orbital effects, usually present in experiments with SETs [23], does not complicate data interpretation. At low B , our data are qualitatively consistent with the predicted universal dependence of the splitting on T_K [10, 11] as well as with the earlier experiment [15] but are incompatible with the universality at high B . The tunable parameters in this work are the energy ϵ_0 of the local orbital measured from the Fermi energy of the leads, and the effective dot-lead coupling $\Gamma = \Gamma_S + \Gamma_D$, where Γ_S and Γ_D are the tunneling rates from the source and drain leads onto the dot. Increasing Γ or $|\epsilon_0 + U/2|$, where U is the quantum dot charging energy,

makes T_K larger [17]. When B is applied, the peak in $G(V_{ds})$ splits into two peaks with the maxima at energies $\pm\Delta_K$ relative to the Fermi energy of the leads. Theory [10, 11] predicts that Δ_K/Δ increases with $\Delta/k_B T_K$, as a universal function, and therefore Δ_K decreases with T_K . Here $\Delta = g\mu_B B$ is the Zeeman splitting of a spin-degenerate orbital level in the absence of Kondo correlations, g is the g-factor and μ_B is the Bohr magneton. We find a marked change in the $\Delta_K(T_K)$ dependence as B increases. At low B , Δ_K decreases with T_K , as predicted [10, 11]. At high B , however, Δ_K increases with T_K when we increase Γ while keeping ϵ_0 constant, but decreases with T_K when we vary ϵ_0 at constant Γ . We conclude that the predicted universality with respect to T_K breaks down at high B .

To measure the differential conductance G , we use standard lock-in techniques with a 17 Hz, 1.9 μ V RMS excitation voltage added to V_{ds} . Depending on the visibility of the peak splitting, we vary the acquisition time of a single V_{ds} scan between 15 minutes and 4 hours with time constants ranging from 1 s to 30 s and, for the smallest splittings that we record, average several measurements. Reproducible splittings were observed several months apart in measurements separated by multiple gate voltage cycles. Our SETs (Fig. 1b, inset) were fabricated on a modulation-doped GaAs/AlGaAs heterostructure containing a two-dimensional electron gas (2DEG) 85 nm below the surface with the sheet density $n_{2D} = 4.8 \times 10^{11} \text{ cm}^{-2}$ and mobility $\mu \geq 5 \times 10^5 \text{ cm}^2/\text{V sec}$. The metal gates (5 nm Ti/ 20 nm Au) were patterned via e-beam lithography followed by lift-off. The tunneling rates Γ_S and Γ_D are controlled by the pairs (V_S, V_T) and (V_S, V_B) of gate voltages. We note that V_S controls both Γ_S and Γ_D . A deviation, $\delta\epsilon$, of the dot energy from the middle of the Coulomb valley is a linear combination of the changes in the gate voltages and V_{ds} :

$$(\delta\epsilon)/e = \alpha_S(\delta V_S) + \alpha_T(\delta V_T) + \alpha_B(\delta V_B) \\ + \alpha_G(\delta V_G) + \alpha_{ds}(\delta V_{ds})$$

where $\alpha_i = C_i/C_{total}$ are the mutual capacitances between the dot and the device gates and leads, expressed as fractions of the total capacitance of the dot. Using standard techniques, we find $U = 1.4 \pm 0.05 \text{ meV}$, $\alpha_{ds} = 0.4 \pm 0.03$, $\alpha_G = 0.024 \pm 0.0018$, $\alpha_S/\alpha_G = 2.4$, $\alpha_T/\alpha_G = 2.5$, and $\alpha_B/\alpha_G = 2.0$. We estimate the actual dot diameter to be $\sim 0.13 \mu\text{m}$, which gives an average orbital level spacing of $\sim 540 \mu\text{eV}$. The external magnetic field B is aligned parallel to the 2DEG plane to $\pm 1^\circ$. Using the spin-flip cotunneling spectroscopy method [14], we obtain the base electron temperature for our dilution refrigerator (Leiden

Cryogenics) $T_{el} \leq 55$ mK and the heterostructure g -factor $|g| = 0.2073 \pm 0.0013$, which gives $\Delta = 12.02 \mu\text{eV/T}$. Fig. 1a shows a Kondo valley between $V_G = -930$ mV and $V_G = -870$ mV flanked by Kondo-free, even-occupied valleys. The Kondo temperature at different gate voltages, obtained from measurements of the temperature dependence of the Kondo conductance [23], is plotted in Fig. 1b. The zero-bias mid-valley Kondo peak (Fig. 1c) is shown for two device configurations: I with $\Gamma=0.53$ meV, $T_K = 0.3$ K, and II with $\Gamma = 0.7$ meV and $T_K = 0.63$ K. We note that, despite the difference in V_G , both I and II correspond to the middle of the Kondo valley, i.e. $\epsilon_0 \sim -U/2$ for both. The choice of the I and II configurations is a compromise between minimizing thermal effects, which favors higher Γ and T_K , and avoiding mixed-valence corrections [18], which limits the largest usable Γ to $|\epsilon_0|/\Gamma \geq 0.5$ [18]. When sweeping between I and II, the relative electron temperature T_{el}/T_K varies from 0.18 to 0.09, and $|\epsilon_0|/\Gamma > 1$ is maintained. To obtain T_K , we fit the temperature dependence $G(T)$ in each configuration to the empirical form $G(T) = G_0[1 + (2^{1/0.22} - 1)(T/T_K)^2]^{-0.22}$ [18]. We estimate Γ independently from the width of the charging peak with the Kondo effect thermally suppressed, and from fits of $T_K(V_G)$ to the Haldane function [17], and find good agreement. Using the height of the Kondo peak to estimate the device asymmetry, Γ_S/Γ_D , we get 24 (I) and 7 (II). To obtain Δ_K from measured $G(V_{ds})$ (Fig. 2a)[23], we first fit the data near each maximum to an analytical function $G^{fit}(V_{ds})$. To account for the slight peak height difference, we subtract a linear background from the fits to equalize the maxima and then obtain V_{ds} for the left and the right peaks by solving $dG^{fit}/dV_{ds} = 0$. Δ_K/e is taken as half the difference between these V_{ds} values. We find that the result varies by no more than $\sim 2 \mu\text{V}$ for different sensible choices of the background slope.

We open the discussion of the results by examining how Δ_K changes with ϵ_0 at constant Γ . Starting with all gate voltages set to I (see Fig. 1, caption), we scan V_G , and then repeat the experiment with II as the starting point. Fig. 3 presents plots of Δ_K as functions of the deviation, δV_G , of V_G from the value that corresponds to I (open squares) and II (filled circles). At all B , Δ_K decreases as V_G is tuned away from the center of the valley, which corresponds to an increase in T_K . This agrees with the earlier observations [15]. Next, we fix the dot energy in the middle of the valley and focus on the changes of the splitting with Γ . A detailed dependence of the mid-valley splitting $\Delta_{K,0}$ on B for the configurations I and II is presented in Fig. 4. First, we note that the lowest magnetic field at which the

Kondo peak shows detectable splitting increases with T_K . Introducing the corresponding Zeeman scale Δ^{onset} , we find $\Delta^{onset} = 0.55 k_B T_K$ (I) and $\Delta^{onset} = 0.4 k_B T_K$ (II). These are in reasonable agreement with the prediction $\Delta^{onset} = 0.5 k_B T_K$ [19] and are somewhat lower than the previously reported $\sim 0.86 k_B T_K$ [15], and $\sim 0.8 k_B T_K$ [13] possibly due to a heavy signal averaging and a lower relative electron temperature ($\sim 1/15$ to $1/6$ in the present work vs $1/6$ in [13], and $1/3$ in [15]). Near the onset, the data show a pronounced suppression $\Delta_{K,0} < \Delta$, consistent with, but much stronger, than in the earlier report by Quay et. al. [13] who used a carbon nanotube-based SET. We note that in the earlier experiments with heterostructure-based SETs [14, 15], $\Delta_{K,0} < \Delta$ was not observed. As Δ increases above $\sim k_B T_K$, $\Delta_{K,0} < \Delta$ is replaced with the $\Delta_{K,0} > \Delta$ regime. To our knowledge, such a transition at a finite B has not yet been reported, although $\Delta_K > \Delta$ was previously observed experimentally [14, 15, 16] [24], found theoretically in the very recent calculations by Hong and Seo [20], and also, for the $B \gg k_B T_K$ regime, predicted by the perturbative method described by Paaske et. al. [22]. At yet higher B , our $\Delta_{K,0}$ data for the more open, higher T_K configuration II exceed those for the lower T_K configuration I. This is opposite of what we find in the fixed Γ experiments (Fig. 3 and its discussion), in which $\Delta_{K,0}$ decreases with T_K regardless of the magnitude of B . To examine the $\Delta_K(T_K)$ dependence at fixed energy in more detail, we follow a constant ϵ_0 , variable Γ path starting in the middle of the Kondo valley (Fig. 5). In the beginning of each sweep, the device is set to I. Then, both V_S and V_G are swept simultaneously so as to keep $\alpha_S(\delta V_S) + \alpha_G(\delta V_G) = 0$, and thus maintain $\epsilon_0 \sim -U/2$. The changes in T_K during such a sweep come from the changes in Γ only, and the device is being tuned continuously from I to II. To verify that the presence of the magnetic field does not reverse the expected increase of Γ with V_S , as may occur, for example, due to an accidental scattering by impurities near the tunnel barriers, we have compared the charging peak widths in configurations I and II measured at zero bias with a 9 Tesla magnetic field applied. We found the ratio of the widths to be 0.6. This is comparable to the ratio of Γ values in I and II at zero magnetic field (0.78), and indicates that the configuration II remains stronger coupled to the leads than configuration I even at the highest available magnetic field. At low B , we observe $\Delta_{K,0} < \Delta$ and $\Delta_{K,0}$ decreasing with increasing V_S (and also Γ and T_K). At fields larger than ~ 4 T, the opposite occurs: $\Delta_{K,0} > \Delta$ and increases as Γ and T_K increase. Thus, at high B , scaling with T_K breaks down: changes of Δ_K with T_K in the constant energy and in the constant Γ experiments

have opposite signs. Interestingly, both the high B and the low B trends shown in Fig. 5 agree qualitatively with the $\Delta_K \rightarrow \Delta$ behavior expected for the limit of small Γ and T_K [10, 11, 21].

In summary, we have measured Δ_K as a function of B and two parameters, ϵ_0 and Γ , that influence T_K . At a sufficiently large B , a crossover occurs to a regime in which a universal dependence of Δ_K on T_K is qualitatively inconsistent with the data. In addition, we observe both $\Delta_K < \Delta$ (low B) and $\Delta_K > \Delta$ (high B) regimes in a single SET system, and find that the transition between the two regimes occurs at B values comparable to those for the crossover.

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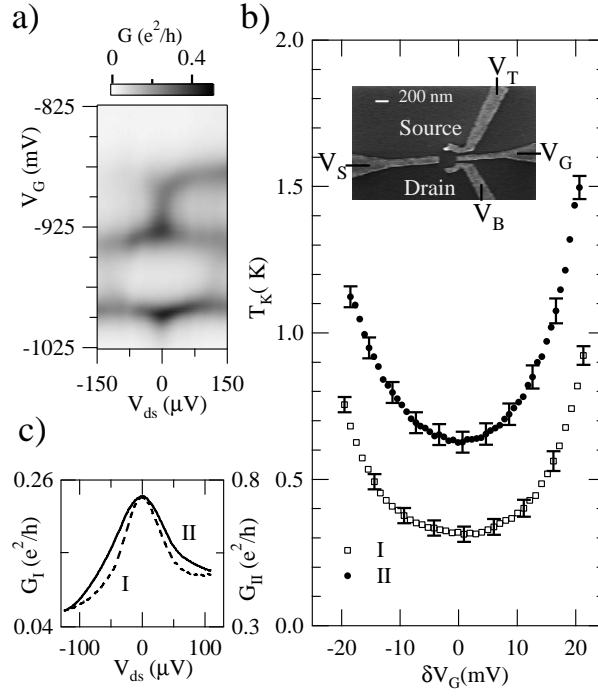


FIG. 1: a) Differential conductance G as a function of V_G and drain-source bias V_{ds} . b) T_K as a function of δV_G for two SET configurations. Inset: An electron micrograph of our SET devices' gate pattern with the gate voltage labeling convention shown. c) The Kondo peak in Configuration I ($V_B = -825$ mV, $V_T = -942$ mV, $V_S = -908$ mV, and $V_G = -914$ mV) and in Configuration II ($V_B = -825$ mV, $V_T = -942$ mV, $V_S = -880$ mV, and $V_G = -984$ mV). The dot occupancy is the same in I and in II.

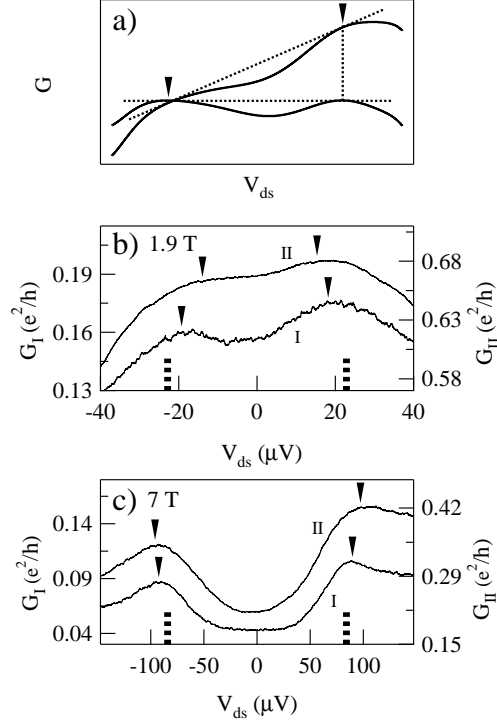


FIG. 2: a) Obtaining Δ_K from measured $G(V_{ds})$. b), c) Representative conductance data in configurations I and II at $B = 1.9$ T (b) and $B = 7$ T (c). The peak positions as determined by our procedure are marked with arrows next to the traces. The Zeeman voltage scale, $V_{ds} = \pm\Delta/e$, is shown with short dashed lines[23].

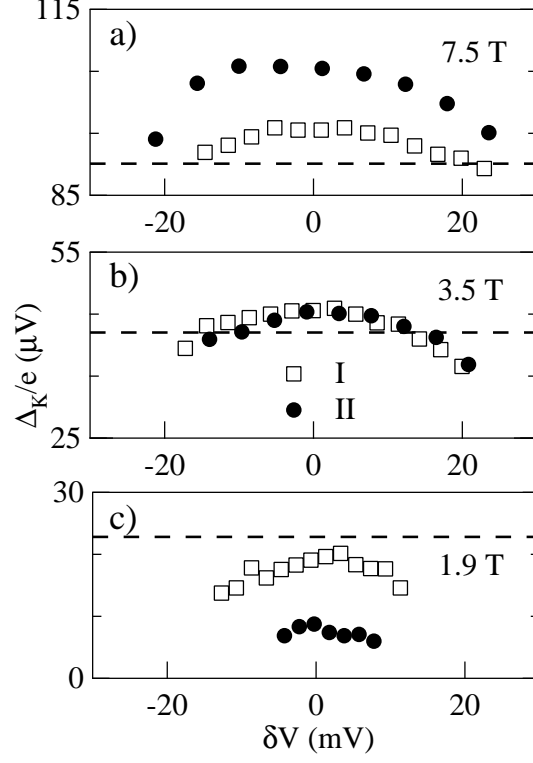


FIG. 3: Variation of Δ_K with gate voltage V_G at different values of B field. $\delta V_G = 0$ corresponds to configuration I (open squares) and II (filled circles) as defined in the caption of Fig. 1. The Zeeman bias voltage scale, Δ/e , for each B is marked with a dashed line. T_K in I and II is 0.3 and 0.63 K, respectively

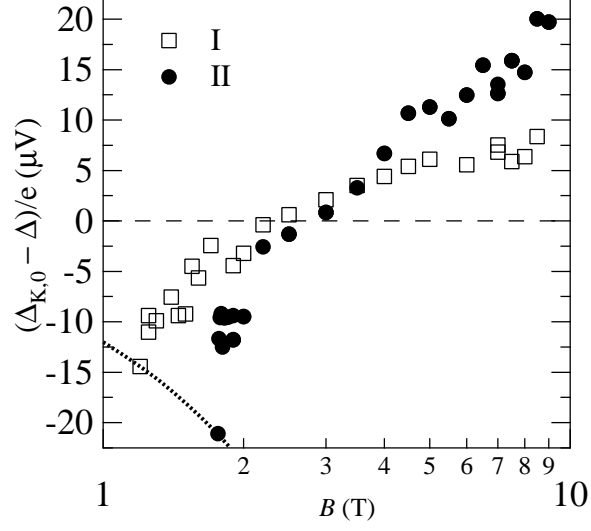


FIG. 4: Deviation of mid-valley splitting $\Delta_{K,0}$ from Zeeman energy Δ as a function of magnetic field. The horizontal dashed line corresponds to $\Delta_{K,0} = \Delta$. The dotted line corresponds to zero peak splitting: $\Delta_{K,0}=0$ [23].

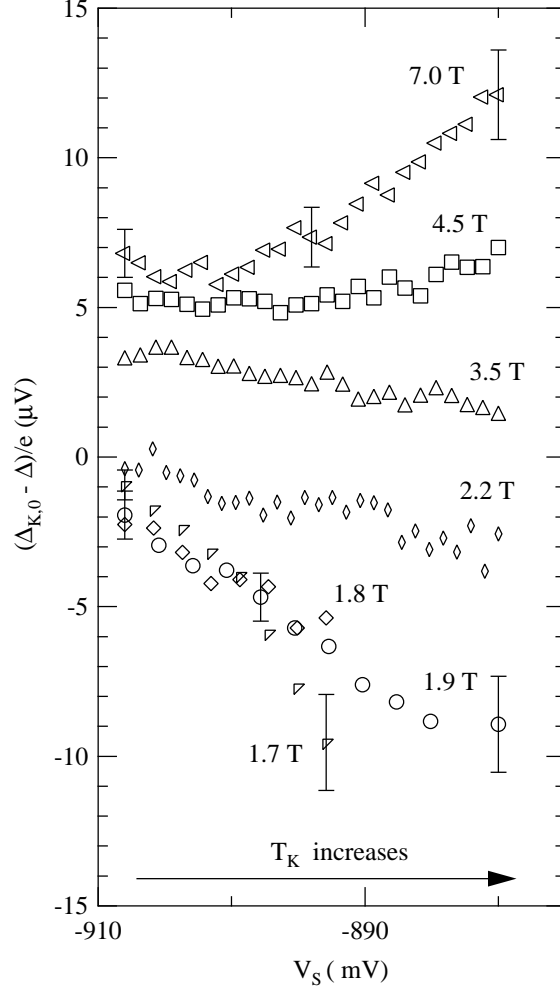


FIG. 5: Deviation $\Delta_{K,0} - \Delta$ as a function of V_S along a constant ϵ_0 path. Both V_S and V_G are swept. The dot energy is set in the middle of the Kondo valley and $\alpha_S(\delta V_S) + \alpha_G(\delta V_G) = 0$ is maintained while V_S is varied.